Development of a fast burn-up method and investigation of transmutation in Generation IV fast reactors

Ph.D. thesis booklet

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Introduction

Generation IV fast reactors are envisaged to operate in closed fuel cycles due to their ability to breed their fuel from $^{238}\text{U}$ or $^{232}\text{Th}$ and burn minor actinides produced by themselves or thermal reactors in the nuclear park [US DoE, 2002]. The production of nuclear waste can therefore be limited to fission products and reprocessing losses, while the reduced waste volume and decay heat can contribute to a more economical use of geological repositories [Salvatores, 2011]. Strategic decisions about the deployment of fast reactors and the transition from open to closed fuel cycle are supported by fuel cycle scenario codes, which are capable of modeling the nuclear fuel cycle along with its most important facilities and the material flows between them [McCarthy, 2009]. The main challenge of scenario studies that concern multiple recycling of the spent fuel is that the evaluation of different fuel cycle strategies requires the tracking of a large number of isotopes in the fuel cycle and the accurate determination of the spent fuel composition.

Due to the high computational cost of detailed burn-up calculations, most scenario codes use burn-up tables or parametrized cross-sections to calculate fuel depletion in the reactors [Boucher, 2010]. Burn-up tables or burn-up dependent cross-section sets, however, may provide inaccurate results if the isotopic composition of the fuel changes greatly, which is the case when multiple recycling of plutonium and minor actinides is considered. Vidal et al. [Vidal, 2006] have introduced the fractions of individual plutonium isotopes and $^{241}\text{Am}$ as cross-section fitting parameters in the CESAR5.3 code, which decreased differences compared to reference calculations by a factor of 6. Leniau et al. [Leniau, 2015] have recently developed a neural-network-based model implemented in the CLASS code to account for changing plutonium compositions in MOX fuel. Their model uses the fractions of $^{238-242}\text{Pu}$ isotopes, $^{241}\text{Am}$, $^{235}\text{U}$ and $^{238}\text{U}$, as well as the irradiation time as input parameters for the neural networks, resulting in less than 3% difference in end-of-cycle inventories obtained with CLASS and MCNP-based MURE depletion calculations.
Objectives

The first aim of the PhD research was to develop a new approximate burn-up method that can provide accurate results in short computational time even in the case of multiple recycling of plutonium and minor actinides. Based on the fitting of one-group cross-sections as functions of the detailed fuel composition (the atomic densities of 15-20 nuclides), I have developed a fast burn-up scheme called FITXS, which I have used to create burn-up models for the Generation IV Gas-cooled Fast Reactor (GFR), Lead-cooled Fast Reactor (LFR) and Sodium-cooled Fast Reactor (SFR), as well as MOX fuel assemblies of Generation III thermal reactors.

The second aim of the thesis was to study minor actinide burning and fissile material breeding properties of the three investigated Generation IV fast reactors. For this purpose, I have analyzed the equilibrium closed fuel cycle operation of the reactors, as well as more complex fuel cycle schemes describing the transition from a Light Water Reactor fleet to a mixed fleet of Generation IV fast reactors and MOX fueled Generation III thermal reactors, and investigated different scenarios concerning the stabilization and reduction of the plutonium and minor actinide inventories.

The results of the fuel cycle studies showed that minor actinide feed results in improved breeding in the three Generation IV fast reactors, which effect was previously observed by Coquelet et al. [Coquelet, 2015] and Meyer et al. [Meyer, 2013] in the SFR. The third aim of the PhD research was to investigate the underlying processes in minor actinide burning and fissile material breeding in the reactors, in particular to identify the reasons behind the increase in breeding gain due to minor actinide feed.

Methods

Three-dimensional core models and fuel assembly models of the reactors were created in the KENO-VI Monte Carlo transport module of the SCALE 6.0 code [ORNL, 2009], which were then used to prepare cross-section databases
Methods

with numerous different fuel compositions in order to perform the least-squares fitting of the one-group cross-sections and the $k_{\text{eff}}$ as second-order polynomial functions of the atomic densities. The burn-up models were verified with burn-up calculations using cross-sections calculated with the SCALE 6.0 code, as well as the fitted cross-sections.

The burn-up models of the Generation IV fast reactors were integrated in equilibrium closed fuel cycle models and transition scenarios using the fuel cycle simulation program JOSSETE (ObJect Oriented Siimulation Program for ScEnario STudiEs), which I have developed for the analyses in C++ programming language. Fuel cycle facilities and models were implemented as classes, and inheritance was used to create the derived classes of specific facility types and calculational methods. The program models facilities and material flows in discrete form, and advances time according to discrete events, which represent the operation of the facilities.

The analysis of the underlying processes in minor actinide burning and fissile material breeding motivated the development of the stochastic models of individual nuclide chains based on discrete-time and continuous-time Markov chains. The models describe the transmutation and decay chains of individual atoms as stochastic processes, and were used to derive closed formulas for finite-time and asymptotic fuel cycle performance parameters, as well as time-dependent transmutation trajectory probabilities, which make it possible to identify the prevailing processes of minor actinide burning and fissile material breeding.

Finally, the results of the closed fuel cycle studies and the Markov chain models of the actinide transmutation chains were applied to analyze minor actinide burning in the three Generation IV fast reactors, including the effect of minor actinide feed on the breeding gain of the reactors. The contributions of different nuclides and transmutation trajectories to the breeding gain were calculated, and the effects of minor actinide feed were analyzed based on sensitivity coefficients and with the use of the Markov chain models in order to identify the reasons behind the improved breeding.
New scientific results

1. I have developed a fast burn-up scheme called FITXS based on the fitting of microscopic one-group cross-sections and the $k_{\text{eff}}$ as functions of the detailed fuel composition, including a wide selection of minor actinide isotopes. I have developed burn-up models with the FITXS scheme for the Generation IV Gas-cooled Fast Reactor, Lead-cooled Fast Reactor and Sodium-cooled Fast Reactor, as well as for MOX fuel assemblies of the Generation III European Pressurized Reactor and VVER-1200, which can determine the spent fuel compositions of the reactors for a wide range of fresh fuel compositions. I have verified the accuracy of the burn-up models using the SCALE 6.0 code [P1, P2, P3].

2. I have developed a fuel cycle simulation program called JOSSETE, with which I have demonstrated the applicability of the FITXS method in fuel cycle simulations and scenario studies by investigating the closed fuel cycles of the three Generation IV fast reactors, taking into account the whole transition from initial state to equilibrium, while the fitting of the $k_{\text{eff}}$ allowed to determine the fresh fuel compositions of the reactors with iteration. Consistently with previous studies in related literature, the results show that the three investigated fast reactors are iso-breeders in the equilibrium due to slight breeding and $^{241}$Pu decay in interim storage, with approximately 1% minor actinide content. Additional minor actinide feed results in improved breeding in the cores [P1, P3, P4].

3. I have shown in scenarios describing the transition from conventional LWRs to a mixed fleet of Generation IV fast reactors and MOX fueled Generation III thermal reactors, that all of the three investigated fast reactors can burn minor actinide stocks that were accumulated in the spent fuels of the conventional LWRs which produced the plutonium for their start-up. I have determined the power ratios of the fast and MOX fueled thermal reactor fleet needed to stabilize or reduce the plutonium inventory, and shown
that fuel composition limits can be met throughout the scenarios if the reprocessed plutonium from spent MOX fuel is recycled in the fast reactors first. A higher power ratio of MOX fueled thermal reactors can counterbalance the improved breeding in the fast reactors due to minor actinide feed in the burner phase, after which a lower thermal reactor power ratio is needed to reach an equilibrium state in the fuel cycle [P5, P6].

4. I have developed the stochastic models of the nuclide transmutation chains based on discrete-time and continuous-time Markov chains. I have shown that the continuous-time Markov chain model can be used to derive both the Bateman equations and time-dependent transmutation trajectory probabilities in the nuclide chains, including decay chains which end in a stable nuclide and actinide transmutation chains which end with fission. I have shown that the transmutation trajectory probability is the general solution of the Bateman equations for linear chains if unit concentration is assumed for the starting nuclide. Transmutation trajectory probabilities in the actinide transmutation chains can be used to identify the prevalent processes in minor actinide burning and fissile material breeding [P7].

5. I have developed a method to count the expected values of labeled transitions in the transmutation chains using the Markov chain models, with which I have derived closed formulas for the calculation of finite-time-integrated and asymptotic fuel cycle performance parameters, such as fission probabilities, average neutron balances, D-factors, the average time until fission and the distribution of fissioned daughter nuclides. Based on the derived formulas I have shown in a simplified closed fuel cycle scheme that the neutron production of the equilibrium fuel integrated for one burn-up cycle equals the asymptotic neutron production of the feed vector [P7, P8].

6. I have investigated the effect of minor actinide feed from spent LWR fuel on the breeding properties of the three Generation IV fast reactors. I have calculated nuclide-wise contributions to the increase in breeding gain
due to minor actinide feed based on sensitivity coefficients, and mapped transmutation trajectories with the highest absolute contributions to the breeding gain based on the Markov chain models. I have shown that the improved breeding in the three fast reactors is mainly due to the production of $^{238}\text{Pu}$ from $^{237}\text{Np}$ and the decreased $^{239}\text{Pu}$ and $^{241}\text{Pu}$ content of the fresh fuel. The improvement is somewhat moderated by the decreased production of $^{239}\text{Pu}$ from $^{238}\text{U}$ and $^{241}\text{Pu}$ from $^{240}\text{Pu}$, as well as the increased $^{238}\text{Pu}$ and $^{245}\text{Cm}$ content. The spectral effects of the increased minor actinide content are much smaller compared to these changes in production and consumption rates [P1, P7].

**List of publications**


References


